

# Spectral properties of correlation functions of fields with arbitrary position dependence in restricted geometries from the ballistic to the diffusive regimes.

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## Abstract

The transition between ballistic and diffusive motion poses difficult problems in several fields of physics. In this work we show how to calculate the spectra of the correlation functions between fields of arbitrary spatial dependence as seen by particles moving through the fields in regions bounded by specularly reflecting walls valid for diffusive and ballistic motion as well as the transition region in between for motion in 2 and 3 dimensions.

Applications to relaxation in nmr are discussed.

## I. INTRODUCTION

The problem of the transition between ballistic and diffusive motion impacts many fields of physics including charge transport in semiconductors, electron transport in mesoscopic systems, fluid flow, heat conduction [1], light transport in random media, [2], [3] relaxation and frequency shifts in magnetic resonance of particles moving in non-uniform fields [4], [5]. In this work we concentrate on the latter problem and present solutions in finite regions in 1, 2 and 3 dimensions valid for ballistic and diffusive motions and the crossover between.

In these problems, the physical effects of interest depend on the spectra of the correlation functions of various field components as seen by the moving particles.

$$S_{1,2}(\omega) = \int_{-\infty}^{\infty} \langle B_1(t) B_2(t+\tau) \rangle e^{-i\omega\tau} d\tau \quad (1)$$

where  $\langle \rangle$  signifies the ensemble average over the gas particles.

As shown first by [6] (see also [7], [8]) and then applied by [9], [10] and [11] these correlation functions are given by the conditional probability  $p(\vec{r}, \tau | \vec{r}_0, 0)$ , the probability that a particle at  $\vec{r}_0$  at  $t = 0$  will be found at a position  $\vec{r}$  at the later time  $\tau$ :

$$R_{1,2}(\tau) = \langle B_1(t) B_2(t+\tau) \rangle = \int \int d^3r d^3r_0 B_1(\vec{r}_0) B_2(\vec{r}) p(\vec{r}, t_0 + \tau | \vec{r}_0, t_0) p(\vec{r}_0, t_0) \quad (2)$$

where  $p(\vec{r}_0, t_0)$  is the probability of finding a particle at  $\vec{r}_0$  at time  $t_0$  which is taken as constant ( $= 1/L$  in 1 dimension). In this paper we consider single-speed transport only.

We treat the motion as a persistent continuous time random walk with isotropic scattering (in 2 and 3 dimensions) and uniform velocity with an exponential distribution of times,  $t$ , between scatterings,

$$\psi(t) = \frac{1}{\tau_c} e^{-t/\tau_c} \quad (3)$$

## II. MOTION IN 1 DIMENSION

Goldstein [12] has shown that in these conditions the conditional probability distribution, considering the motion as the continuum limit of a persistent random walk, satisfies the telegrapher's equation in one dimension. As shown by Masoliver et al [13] the equation can be solved by separation of variables and the conditional probability with reflecting boundary conditions at  $x = 0, L$  is given by

$$p(x, t | x_0, 0) = \frac{1}{L} \left\{ \sum_n \cos\left(\frac{n\pi x}{L}\right) \cos\left(\frac{n\pi x_0}{L}\right) \right\} \left\{ \cosh\left(\frac{s_n t}{2\tau_c}\right) + \frac{1}{s_n} \sinh\left(\frac{s_n t}{2\tau_c}\right) \right\} e^{-\frac{t}{2\tau_c}} \quad (4)$$

with  $s_n = \sqrt{1 - 4\omega_n^2 \tau_c^2}$  and  $\omega_n = n\pi v/L$  with  $v$  the velocity of the particles. Note the change in behavior as  $s_n$  goes from real to imaginary. The behavior of this solution is discussed in some detail in [13] and [14]. There is a delta function peak leaving the source with velocity,  $v$ . This peak diminishes in time as particles are scattered and a wake of scattered particles builds up behind the peak. The motion can be followed through successive wall reflections.

Then according to (2) the field correlation function is given by

$$R_{1,2}(\tau) = \sum_n F_1(k_n) F_2(k_n) \left\{ \cosh\left(\frac{s_n \tau}{2\tau_c}\right) + \frac{1}{s_n} \sinh\left(\frac{s_n \tau}{2\tau_c}\right) \right\} e^{-\frac{\tau}{2\tau_c}} \quad (5)$$

with

$$F_{1,2}(k_n) = \frac{1}{L} \int_0^L dx B_{1,2}(x) \cos(k_n x) \quad (6)$$

and

$$k_n = n\pi/L \quad (7)$$

The Fourier transform of this gives the desired spectrum which determines physical phenomena.

$$S_{1,2}(\omega) = \sum_n F_1(k_n) F_2(k_n) \left( \frac{2\omega_n^2 \tau_c}{\omega^2 + [(\omega^2 - \omega_n^2)^2 \tau_c^2]} \right) \quad (8)$$

For not too high  $\omega$ :  $\omega \ll \pi v/L$  this result is consistent with one obtained earlier for the diffusion regime of motion [10], [11].

This result is consistent with calculations of the position- correlation function,  $B_1 = B_2 \sim x$  for various values of normalized mean free path,  $l' = v\tau_c/L$  presented in [5], see figure 2 in that paper. Notice as the motions approaches ballistic,  $l' \gg 1$ , the relaxation shows a resonance behavior when a harmonic of the wall collision frequency coincides with the Larmor frequency, which has not been noted previously. The resonance peaks are smoothed by averaging over the velocity distribution. Experiments to observe this are under way.

It may be shown that for a perturbation field with a uniform gradient  $B = Gx$  the series (6) can be summed in closed form:

$$S = 2 \left( \frac{G}{\omega} \right)^2 \frac{v^2 \tau_c}{(1 + \omega \tau_c)^2} F(\omega) \quad (9)$$

where

$$F(\omega) = 1 - \text{Im} \left[ (\omega \tau_c + i) \frac{\tan \left( \frac{\omega \tau_b}{2} \sqrt{1 - i/\omega \tau_c} \right)}{\left( \frac{\omega \tau_b}{2} \sqrt{1 - i/\omega \tau_c} \right)} \right] \quad (10)$$

$$\tau_b = \frac{L}{v} \quad \text{characteristic ballistic time}$$

In the limit  $\omega \tau_c \ll 1$  (9) goes over to the diffusion theory result, see [9].

### III. MOTION IN 2 DIMENSIONS.

#### A. Solution in free space

The obvious extension of the above ideas is to apply them to higher dimensions by writing the Telegrapher's equation (TE) with the appropriate form of  $\nabla^2$ . This was first suggested by Mark Kac, [16]. However the solution of the TE cannot represent a conditional probability in two or higher dimensions as it can take on negative values, [17]. Morse and Feshbach [18] discuss the interesting properties of solutions of the TE (as well the wave equation) in 2 dimensions. This point seems to have been missed in an otherwise interesting and useful work [19], which nonetheless finds the same solution for the spectrum of the conditional probability function as given by [13]. In a remarkable paper Masoliver et al [20] have shown that the conditional probability function for a persistent random walk in two dimensions, with the time between scattering distributed according to (3) and a uniform distribution of scattering angles (s-wave scattering) for particles starting at the origin of coordinates with velocity  $v$ , satisfies the 2 D TE with an additional source term

$$\rho(r, t) = \frac{v^2}{2\pi r} e^{-t/\tau_c} \frac{\partial}{\partial r} \left( \frac{\delta(r - vt)}{r} \right) \quad (11)$$

The source moves along with the unscattered particles and represents the scattered particles that are not accounted for in the homogeneous TE. The authors give the solution for motion in the infinite domain as:

$$P_{02}(r, t) = e^{-t/\tau_c} \left[ \frac{\delta(r - vt)}{2\pi r} + \frac{1}{2\pi v \tau_c \sqrt{(vt)^2 - r^2}} e^{\frac{\sqrt{(vt)^2 - r^2}}{v \tau_c}} \Theta(vt - r) \right] \quad (12)$$

where  $\Theta$  is the unit step function. The authors also give the Fourier-Laplace transform of the solution

$$\begin{aligned} \hat{P}_{02}(\vec{Q}, s = i\omega) &= \int d^2r \int_0^\infty p(r, t) e^{(i\vec{Q} \cdot \vec{r} - st)} dt \\ &= \frac{\tau_c}{\left[ (1 + i\omega \tau_c)^2 + v^2 \tau_c^2 |\vec{Q}|^2 \right]^{1/2} - 1} \end{aligned} \quad (13)$$

The expression (13) and the equivalent one for three dimensions have been rederived by Kolesnik, [21], who gives a general treatment applicable to an arbitrary number of dimensions.

## B. Solution in bounded rectangular region.

We now extend these results to find the spectrum of the field correlation function in a rectangular region bounded by reflecting walls. This solution is presented here for the first time. As we desire the spectrum of a field correlation function, we will work with the Fourier transform of  $P_{02}(\vec{x}, \tau)$ , where we are using the 0 subscript to denote the free space solution,

$$P_{02}(\vec{x}, \tau) = \frac{1}{(2\pi)^3} \int d^2Q \int d\omega \tilde{P}_0(\vec{Q}, \omega) e^{-i(\vec{Q} \cdot \vec{x} - \omega\tau)} \quad (14)$$

where  $\tilde{P}_{02}(\vec{Q}, \omega) = \hat{P}_{02}(\vec{Q}, s = i\omega)$  (13)

We use the method of images to find the conditional probability function in the presence of the rectangular boundaries at  $x = \pm L_x/2$ ,  $y = \pm L_y/2$ . The location of the images is sketched in fig.1. Wall reflections are taken into account by considering particles coming from the image sources. The desired conditional probability function for a restricted rectangular domain (with specularly reflecting walls) is given by the superposition of probability coming from the original source and all the images.

We see that the physical region is repeated periodically throughout the plane and there is one image point in each cell (the positions of the images are not the same in every cell). Then we can write the solution as

$$P(\vec{r}, \tau | \vec{r}_0, 0) = \sum_i P_{02}(\vec{r} - \vec{r}_{0,i}, \tau) \quad (15)$$

where  $\vec{r}_{0,i}$  is the location of the  $i^{th}$  image. ( $i = 0$  denotes the physical source).

Fig. 2A shows the behavior of  $P_{02}(\vec{r}, \tau)$ , (15) for the parameters,  $v = 1$ ,  $L = 2$ ,  $\lambda = 0.2$  and  $t = 2.7t_b$ , where  $t_b = L/2v$ . Figure 2B show a cross section along the x axis of the function shown in fig.2A.

Then (2)

$$R_{1,2}(\tau) = \langle B_1(t) B_2(t + \tau) \rangle = \int \int d^2r d^2r_0 B_1(\vec{r}_0) B_2(\vec{r}) \sum_i P_{02}(\vec{r} - \vec{r}_{0,i}, \tau) p(\vec{r}_{0,i}, t_0) \quad (16)$$

where the integrals are taken over the physical cell,  $(p(\vec{r}_{0,i}, t_0) = \frac{1}{L_x L_y})$ .

Integrating over  $d^2r_0$  in the physical cell means that each image point in each cell will cover its entire cell and the sum over images and integration over the physical cell can be calculated by taking the function  $P_{02}$  to be the infinite domain function integrated over all space if we continue the field periodically as explained in fig.1. This idea was introduced by Wayne and Cotts [6] (see also [7])

We can rewrite (16) as

$$R_{1,2}(\tau) = \frac{1}{L_x L_y} \int_{-L_x/2}^{L_x/2} \int_{-L_y/2}^{L_y/2} dx dy \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dx_0 dy_0 \tilde{B}_1(\vec{r}_0) B_2(\vec{r}) P_{02}(\vec{r} - \vec{r}_0, \tau) \quad (17)$$

where  $\tilde{B}_1(\vec{r}_0)$  is the periodic extension of  $B_1(\vec{r}_0)$  beyond the physical cell.

Writing  $P_{02}(\vec{r} - \vec{r}_0, \tau)$  in terms of its Fourier transform (14) we find for the spectrum of the correlation function ( $l_x, l_y$  are integers)

$$S_{1,2}(\omega) = \frac{1}{L_x L_y} \sum_{l_x, l_y} \beta_{l_x, l_y} B_2(\vec{q}_{l_x, l_y}) \tilde{P}_{02}(\vec{q}_{l_x, l_y}, \omega) \quad (18)$$

with  $\vec{q}_{l_x, l_y} = \left(\frac{l_x \pi}{L_x}, \frac{l_y \pi}{L_y}\right)$ ,  $\beta_{l_x, l_y} = \frac{1}{L_x L_y} \int_{-L_x/2}^{L_x/2} \int_{-L_y/2}^{L_y/2} dx_0 dy_0 e^{i\vec{q}_{l_x, l_y} \cdot \vec{x}_0} B_1(\vec{x}_0)$  where the integral is taken over one period and

$$B_2(\vec{q}_{l_x, l_y}) = \int_{-L_x/2}^{L_x/2} \int_{-L_y/2}^{L_y/2} dx dy B_2(\vec{x}) e^{-i(\vec{q}_{l_x, l_y} \cdot \vec{x})}.$$



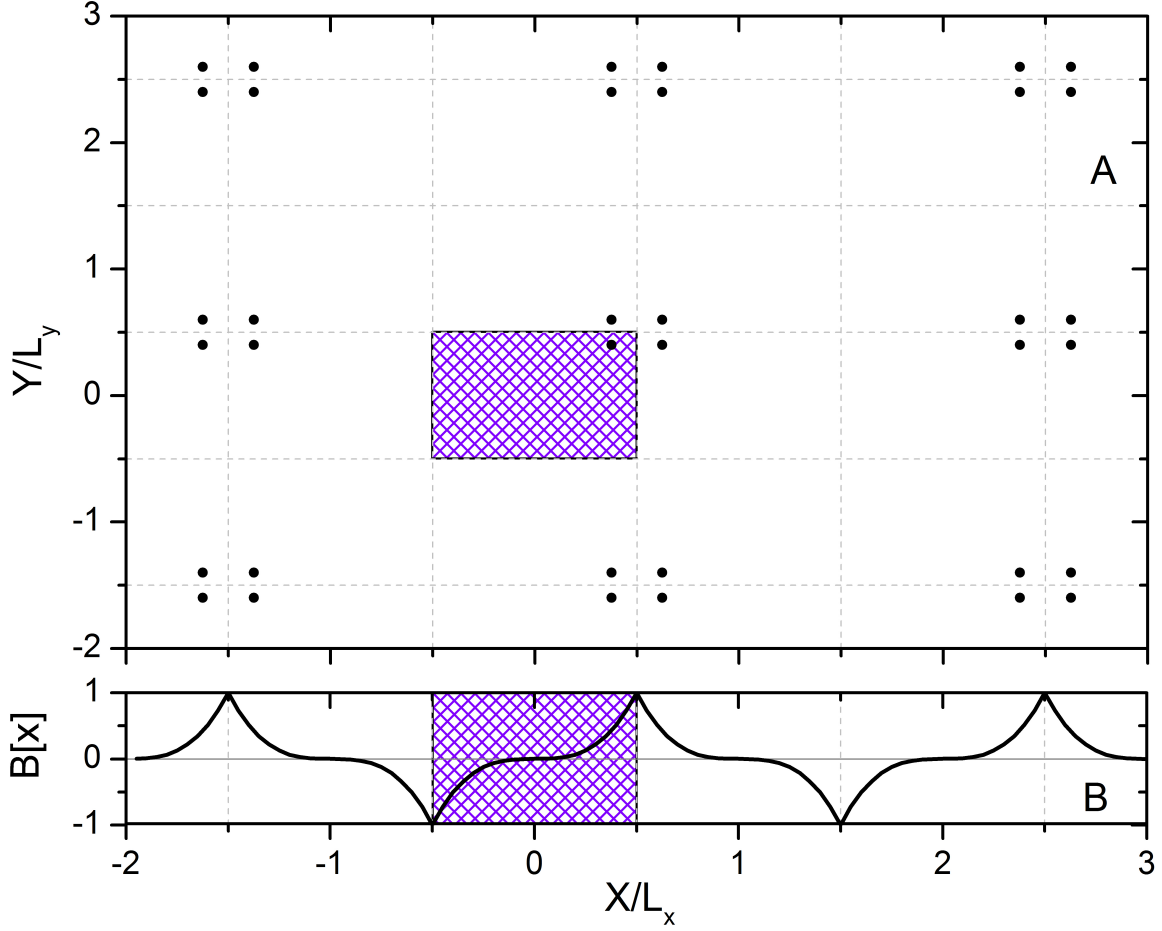


FIG. 1. A) Showing the physical region (shaded) and a few of the periodic repetition cells. A source point and its images are shown. We consider the conditional probability function as a wave travelling from the source to the observation point, P. It is useful to use reciprocity and consider the wave as travelling from P to the source. The solution is then given by considering the wave as travelling in an unrestricted domain and taking the sum of all the waves reaching all the image points. In travelling from the physical source to the wall at  $x=L/2$  in the physical case with boundaries, the particles see  $x$  increasing to the right. Trajectories in this region are replaced by trajectories in the region between the image point and the wall so in this region the effective value of  $x=x$  must be seen to be increasing going from the image to the wall. Thus  $x$  has to be taken as periodic with period  $2L$ . An arbitrary field depending on position must be treated in the same way, (as shown in B) which is a plot of an arbitrary field varying with  $x$  vs.  $x$  (solid line)), and hence must be taken as periodic.

#### IV. SOLUTION IN 3 DIMENSIONS

##### A. Solution in free space

Masoliver et al [20] (see also [22]) give the form of the Fourier Laplace transform of the conditional probability function in three dimensions as

$$\hat{P}_{03}(\vec{Q}, s) = \int d^3r \int_0^\infty p(r, t) e^{(i\vec{Q} \cdot \vec{r} - st)} dt = \tau_c \frac{\tan^{-1}\left(\frac{Qv\tau_c}{1+s\tau_c}\right)}{\left(Qv\tau_c - \tan^{-1}\left(\frac{Qv\tau_c}{1+s\tau_c}\right)\right)} \quad (19)$$

The same result was later obtained by [21] using a different method valid for any number of dimensions.

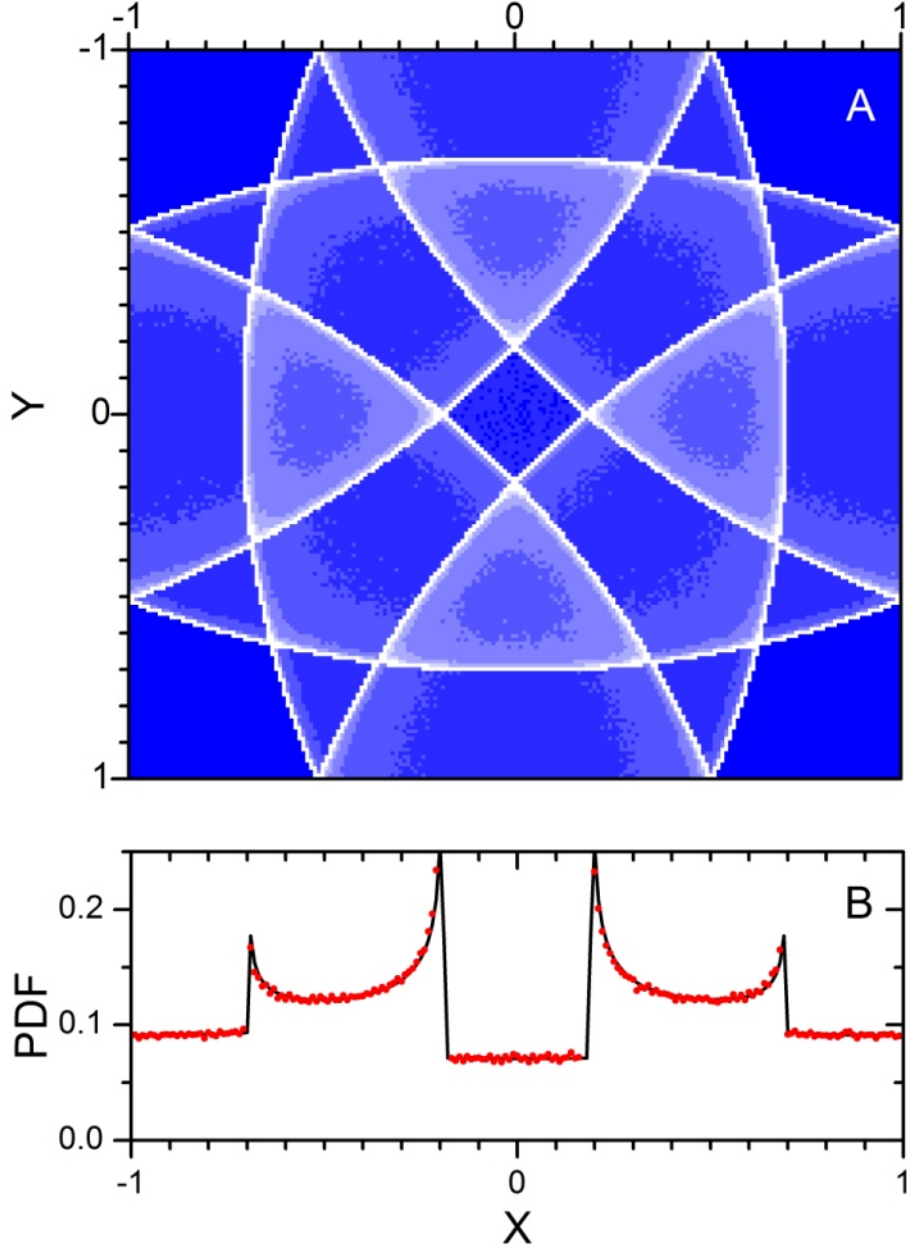


FIG. 2. A) Showing a snapshot of  $P(\vec{r}, \tau)$  for parameters given in the text. The white lines are the peak of unscattered particles while the brightness of the contrast indicates the build up of the wake of scattered particles. B) A cross section along the  $x$  axis of the function  $P_{02}(\vec{r}, \tau = 2.7)$  shown in A). The smaller peaks at  $\pm 0.7$  are the particles that made a collision with the walls at  $x = \pm 1$ . The larger peaks represent the crossing of the two "waves" emanating from the image points at  $(\pm 2, \pm 2)$  and are larger because they are the sum of the two "waves". Solid lines are the result of equation (15) while the dots are the results of a Monte-Carlo simulation.

### B. Solution in a rectangular box

The same reasoning applies here as in 2 dimensions above, the crucial point being that there is one image point per cell.

The spectrum of the correlation function is then:

$$S_{1,2}(\omega) = \sum_{l_x, l_y, l_z} \beta_{l_x, l_y, l_z} B_2(\vec{q}_{l_x, l_y, l_z}) \tilde{P}_{03}(\vec{q}_{l_x, l_y, l_z}, s = i\omega) \quad (20)$$

where  $\vec{q}_{l_x, l_y, l_z} = \left(\frac{l_x \pi}{L_x}, \frac{l_y \pi}{L_y}, \frac{l_z \pi}{L_z}\right)$ ,

$$\beta_{l_x, l_y, l_z} = \frac{1}{V} \int_{-L_x/2}^{3L_x/2} \int_{-L_y/2}^{3L_y/2} \int_{-L_z/2}^{3L_z/2} dx_0 dy_0 dz_0 e^{i \vec{q}_{l_x, l_y, l_z} \cdot \vec{x}_o} \tilde{B}_1(\vec{x}_o) \quad (21)$$

(each integration is over one complete period) and

$$B_2(\vec{q}_{l_x, l_y, l_z}) = \frac{1}{V} \int_{-L_x/2}^{L_x/2} \int_{-L_y/2}^{L_y/2} \int_{-L_z/2}^{L_z/2} dx dy dz B_2(\vec{x}) e^{-i(\vec{q}_{l_x, l_y, l_z} \cdot \vec{x})} \quad (22)$$

## V. APPLICATIONS

In the following we show some illustrative examples of the use of the above technique.

### A. Position auto-correlation functions (Uniform gradient field)

In Fig. 3A) we show the calculation for the spectrum of the  $x-x$  auto-correlation function, determining relaxation times for a field with uniform gradient, as is usually assumed in studies of relaxation, for the one dimensional case for various values of damping using (9).

The results for 2 and 3 dimensions are:

2 dimensions:

$$S_{xx}^{(2D)}(\omega) = \frac{8}{\pi^4} L_x^2 \sum_{n=odd} \frac{1}{n^4} \text{Re} \left[ \hat{P}_{02}(q_n, s = i\omega) \right] \quad (23)$$

3 dimensions:

$$S_{xx}^{(3D)}(\omega) = \frac{8}{\pi^4} L_x^2 \sum_{n=odd} \frac{1}{n^4} \text{Re} \left[ \hat{P}_{03}(\vec{q}_n, s = i\omega) \right] \quad (24)$$

$$\vec{q}_n = \left[ \frac{n_x \pi}{L_x}, \frac{n_y \pi}{L_y}, \frac{n_z \pi}{L_z} \right] \quad (25)$$

For large  $\tau_c$  ( $\lambda \gg L$ ), we see that there is a series of resonances, which will be broadened by averaging over a realistic velocity distribution, and non-specular wall reflections. These are Rabi resonances which occur when a harmonic of the periodic motion coincides with the Larmor frequency. Similar resonances have been noted in cylindrical geometry in another context [24]. For  $\tau_c = 1$  the resonances are still slightly visible, while at  $\tau_c = 0.1$  we have approached the diffusion behavior. In 2 dimensions the resonances are less peaked because closed orbits are less probable. In 3D the peaks (even in the undamped case) are reduced to steps.

In the diffusion limit (green curves) we see the usual behavior at low frequencies approaching a constant as  $\omega \rightarrow 0$  (non-adiabatic region). For higher frequencies the spectrum starts to decrease as  $(1/\omega^2)$ , (adiabatic regime) and at still higher frequencies ( $\omega \tau_c \gg 1$ ) it falls as  $(1/\omega^4)$ , the super-adiabatic region [23]. For moderate (blue) and light (red) damping, the  $1/\omega^2$  behavior disappears and we go directly from a constant to  $1/\omega^4$  behavior.

### B. Short range interaction between the walls and the spin of the particles

This problem has been discussed in [10] using the conditional probability density from diffusion theory. Here in figure 4 we present the results valid for the diffusion and ballistic cases and the transition in between calculated using the 2 dimensional conditional probability (18) taking the interaction to be represented by a effective magnetic field:

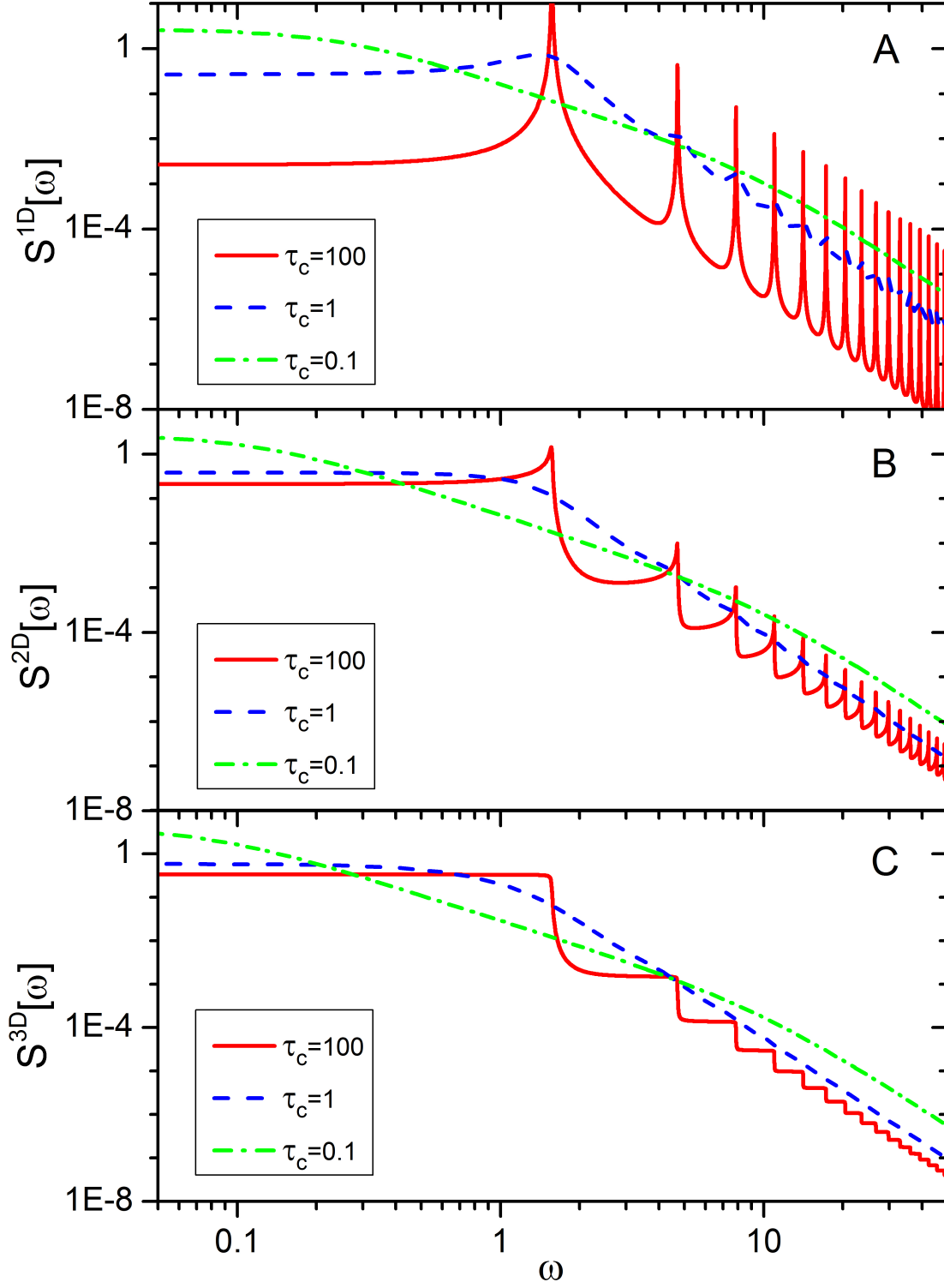


FIG. 3. Showing the spectrum of the autocorrelation function of position  $x$  for various values of the collision time,  $\tau_c$ , vs. dimensionless frequency  $\omega\tau_b$ . A) For one dimension, B) The same for a square in 2 dimensions, C) For a cube in three dimensions

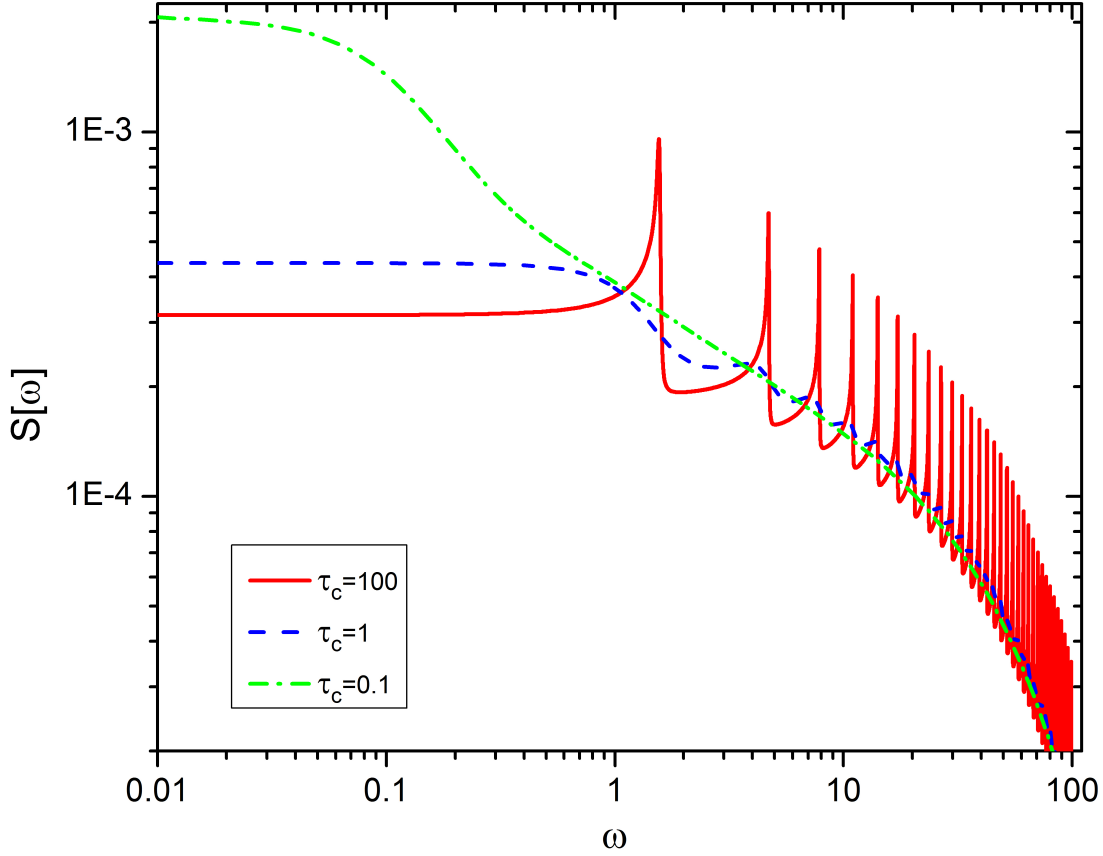


FIG. 4. Results of the auto-correlation function of the field given in equation (26) for the range of force,  $\lambda = .005$ , normalized to the size of the box and various values of  $\tau_c$ .

$$B_{eff}(x) = b_a \left( e^{-(L/2+x)/\lambda} - e^{-(L/2-x)/\lambda} \right) \quad (26)$$

where the range of the interaction is  $\lambda$ .

Here we see the influence of the shape of the field on the spectrum.

In the medium and underdamped cases the constant behavior goes over, as frequency increases, to a  $(1/\sqrt{\omega})$  behavior before reaching the super-adiabatic region  $(1/\omega^4)$ . The extent of the  $(1/\sqrt{\omega})$  behavior will become larger as the range of the force decreases.

In contrast to the spectrum of the position auto-correlation function, in the case of a limited range force the higher frequency results are independent of the damping.

## VI. CONCLUSIONS

We have found expressions for the spectrum of the correlation function of a pair of fields as seen by particles executing a continuous time, persistent random walk, with exponential distribution of flight times in rectangular boundaries with specular reflection and single velocity for 2 and 3 dimensions. The results are valid for all values

of the damping parameter  $\lambda/L = \tau_c/\tau_b$ , allowing calculations in the ballistic and diffusive regimes as well as the transition region in between.

These conditional probability spectra allow the calculation of the spectrum of the correlation functions of fields with arbitrary position dependence and thus the study of relaxation and frequency shifts in arbitrary fields. The results are valid as long as the trajectories are not influenced by the fields.

It is expected that the results will find applications in other fields such as transport in mesoscopic systems.

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## VIII. REFERENCES

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- [1] Maris, H.J. Rev. Mod. Phys. **49**, 341 (1977)
  - [2] Elaloufi, R., Carminati, R. and Greffer, J.J., J. Opt. Soc. Am. **21**, 1430 (2004)
  - [3] Durian, D.J. and Rudnick, J., J. Opt. Soc. Am. **14**, 235 (1977)
  - [4] Cates, G.D., Schaefer, S.R. and Happer, W., Phys. Rev. **A37**, 2877 (1988)
  - [5] Golub, R., Rohm, R.M. and Swank, C.M., Phys. Rev. **A 83**, 023402 (2011)
  - [6] Wayne, R.C. and Cotts, R.M., Phys. Rev. **151**, 264, (1966)
  - [7] Tarczón, J.C. and Halperin, W.P. Phys. Rev. **D32**, 2798, (1985)
  - [8] Oppenheim, I. and Bloom, M., Can J. of Physics **39**, 1961)
  - [9] McGregor, D.D. Phys. Rev. **A41**, 2631 (1990)
  - [10] Petukhov, A.R., Pignol, G., Jullien, D. and Andersen, K.H. Phys. Rev. Letts. **105**, 170401 (2010)
  - [11] Clayton, S.M., Journal of Magnetic Resonance **211** 89–95 (2011)
  - [12] Goldstein, S. Q. J. Mech. Appl. Math. **IV**, 129 (1951)
  - [13] Masoliver, J., Porrá, J.M. and Weiss, G.H., Phys Rev **E48**, 939 (1993)
  - [14] Golub, R. and Swank, C.M., arXiv:1012.4006 (2010)
  - [15] McGregor, D.D., Phys. Rev. **A41**, 2631, (1989)
  - [16] Kac, M., Rocky Mountain Journal of Mathematics, **4**, Number 3, 497 (1974)
  - [17] Porrá, J.M., Masoliver, J. and Weiss, G.H., Phys. Rev. **E55**, 7771 (1997)
  - [18] Morse, P.M. and Feshbach, H. '*Methods of Theoretical Physics*', McGraw-Hill, New York, 1953
  - [19] Kolesnik, A.D. and Pinsky, M.A., J Stat. Phys. **142**, 828 (2011)
  - [20] Masoliver, J., Porrá, J.M. and Weiss, G.H., Physica **A193**, 469 (1993)
  - [21] Kolesnik, A.D., J.Stat. Phys **131**, 1039 (2008)
  - [22] Claes, I. and Van der Broeck, C. J Stat. Phys. **49**, 383 (1987)
  - [23] Scherer, L.D. and Walters, G.K., Phys Rev. **139**, 1398 (1965)
  - [24] Pendlebury, J.M. et al, Phys. Rev **A 70**, 032102 (2004) and Barabanov, et al, Phys Rev **A74**, 052115 (2006)